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LeA 34,849

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPLICANT: Christian Wambrecht et al.
SERIAL NO: 10/092,077
FILED: March 6, 2002
TITLE: NEW POLYURETHANES AND THEIR
USE FOR THE THICKENING OF
AQUEOUS SYSTEMS

DECLARATION

I, Frank Sauer, a citizen of Germany, residing Querstr. 20, D-40764 Langenfeld, Germany, declare as follows:

1. That I have the following technical education and experience:
That I have studied at the University of Cologne (Köln) and received a doctor's degree in natural science at the University of Cologne (Köln) in 1993.

That since October 1999 I have been employed with Borchers GmbH, one of the assignees of the above-identified application, as a research chemist in the field of the chemistry of polyurethanes and have mean-while more than 5 year's experience in that field.

That I am a co-inventor of the above-identified application and familiar with the Office Action dated June 10, 2004.

2. That for overcoming the Rejection the following tests have been made under my supervision:

A) Comparative examples with and without Isophorone diamine (IPDA)

The polymer mentioned in our application was synthesized both with and without the use of Isophorone diamine as a chain-lengthening agent and examined the rheological properties of the resulting polymers:

	Test 1 Quantity in [g]	Test 2 Quantity in [g]	
Polyether VP LS 20055	100.00	100.00	
Octanol	1.60	1.60	
Decanol	2.50	2.50	
Isophorone diisocyanate (IPDI)	6.75	6.75	
Isophorone diamine (IPDA)	0.20	-	
Dibutyltindilaurate (DBTL = catalyst)	0.05	0.05	
Total quantity of PU	111.10	110.90	
Weight of active compound	111.10	110.90	25.0 %
RPDE	44.44	44.36	10.0 %
Levalin FD	55.55	55.45	12.5 %
Emulgator WN	55.55	55.45	12.5 %
Water	177.76	177.44	40.0 %
Total formulation containing 25 % PU	444.40	443.60	100.0 %

Method:

The polyether is dehydrated for two hours at 115°C with a stream of N₂ or vacuum (Karl-Fischer analysis, required value = < 200 ppm). RPDE is added and the two alcohols, IPDI and DBTL are added successively at 115°C.

Test 1:

The mixture is allowed to react at 115°C so long as a clearly visible NCO band can still be identified via a IR-spectrometer (IR control every 15 mins). IPDA is added and the mixture stirred once again for 30 mins.

Test 2:

The mixture is stirred at 115°C until NCO = 0 (for about 2-2.5h).

The both formulations are then prepared as mentioned above, with Levalin- FD, Emulgator WN and water.

Test 1 is according to our invention; Test 2 is a comparative example.

Testing of the rheological properties:

Testing of the thickeners according to test 1 and test 2 in a standard lacquer based on

Neocryl XK 62: 70.0 % Neocryl XK62
 27.9 % water
 2.0 % Butoxyethanol (BG)
 0.1 % Byk 36
 Total: 100.0 %

2 g of each of the thickener from test 1 and test 2 are each introduced into 98 g of the standard lacquer and dispersed for 15 minutes. The mixtures are left to stand for 24 hours and their rheological properties determined (Haake RS1).

Results of the rheological analysis:

see Appendix 1

Comparative tests with Borch Gel VP 0024			
Rheological effectiveness	Test 1 (with IPDA)	Test 2 (without IPDA)	Efficiency
Shear rate 10.3 s^{-1}	836 mPas	700 mPas	+ 19.4% (V1)
Shear rate 10000 s^{-1}	126 mPas	116 mPas	+ 8.6% (V1)

The mixture using IPDA according to our invention (Test 1) clearly has a higher rheological efficiency than the mixture without the diamine (IPDA), both in the "high shear range" (+ 8.6 %) and in the "low-shear range" (+ 19.4 %)

B) Comparative examples with US 4,079,028

A formulation from the examples in the Emmons patent (US 4,079,028) was reworked using a monoalcohol (Test 3), a monoamine (Test 4) and a combination of a monoalcohol and a diamine (Test 5). Test 5 is an example according to our invention, Test 3 and 4 are comparative examples according to the teaching of the Emmons patent (US 4,079,028).

Basis for the comparative examples are the examples 18-74T in the Emmons patent (US 4,079,028: column 16/examples 18-74T) with the title: "Isocyanate terminated prepolymers of polyoxyethylene glycol and diisocyanate capped with aliphatic alcohols or amines".

In particular the example 74M was used as a comparison (of Column 17/ Table 2) and correspondingly modified:

a) Test 3

Polymer components		Raw materials	Molecular weight	Test 3	Eq. (val)
PEG (4.000 - 20.000 g/mol)		PEG 8000 g/mol (difunctional)	8000.0 g/mol	100 g	0.025
Monoalcohol		Dodecanol	186.3 g/mol	4.65 g	0.025
Monoamine		Dodecylamine	185.4 g/mol	-	-
Diamine		Isophorone diamine (IPDA)	170.3 g/mol	-	-
Diisocyanate		Hexamethylene diisocyanate (HDI)	168.2 g/mol	4.20 g	0.050
Catalyst		Dibutyltin dilaurate (DBTL)		0.05 g	-
			[NCO] : [OH, NH]		1 : 1

b) Test 4

Polymer components		Row materials	Molecular weight	Test 4	Eq. (val)
PEG (4.000 - 20.000 g/mol)		PEG 8000 g/mol (difunctional)	8000.0 g/mol	100 g	0.025
Monoalcohol		Dodecanol	186.3 g/mol	-	-
Monoamine		Dodecylamine	185.4 g/mol	4.63 g	0.025
Diamine		Isophorone diamine (IPDA)	170.3 g/mol	-	-
Diisocyanate		Hexamethylene diisocyanate (HDI)	168.2 g/mol	4.20 g	0.050
Catalyst		Dibutyltin dilaurate (DBTL)		0.05 g	-
			[NCO] : [OH, NH]		1 : 1

c) Test 5

Polymer components		Row materials	Molecular weight	Test 5	Eq. (val)
PEG (4.000 - 20.000 g/mol)		PEG 8000 g/mol (difunctional)	8000.0 g/mol	100 g	0.025
Monoalcohol		Dodecanol	186.3 g/mol	4.65 g	0.025
Monoamine		Dodecylamine	185.4 g/mol	-	-
Diamine		Isophorone diamine (IPDA)	170.3 g/mol	0.50 g	0.006
Diisocyanate		Hexamethylene diisocyanate (HDI)	168.2 g/mol	4.70 g	0.056
Catalyst		Dibutyltin dilaurate (DBTL)		0.05 g	-
			[NCO] : [OH, NH]		1 : 1

Method:

The polyether is dehydrated for two hours at 115°C with a stream of N₂ or vacuum (Karl-Fischer analysis, required value: ≤ 200 ppm). Then dodecanol (Test 3) or dodecylamine (Test 4) or the combination of dodecanol and IPDA (Test 5) and then the diisocyanate (HDI) and the catalyst DBTL are added successively at 115 °C. Then every mixture is stirred at 115°C until the NCO=0 (IR-control, depending on the mixture about 30 mins to 1 hour). Then the polymer is formulated in an amount of 20% in 80 % water.

Testing of the rheological properties:

Testing in a standard lacquer based on

Neocryl XK 62: 70.0 % Neocryl XK62
 27.9 % water
 2.0 % Butoxyethanol (BG)
 0.1 % Byk 36
Total: 100 %

4 g of each of the thickeners from test 3, test 4 and test 5 are each introduced into 96 g of the standard lacquer and dispersed for 15 minutes. The mixtures are left to stand for 24 hours and their rheological properties determined (Haake RS1)

Results of the rheological analysis:

see Appendix 2:

Test 3 with Dodecanol; Test 4 with Dodecylamine; Test 5 with Dodecanol and IPDA

Comparative tests with Emmons examples based on test 74M			
Rheological effectiveness	Test 3	Test 4	Test 5
Shear rate 10.3 s ⁻¹	714 mPas	81 mPas	1837 mPas
Shear rate 10000 s ⁻¹	65 mPas	36 mPas	133 mPas

Conclusion:

The use of a diamine according to our invention (Test 1 and 5) such as Isophorone diamine improves the rheological efficiency in all cases.

- I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or

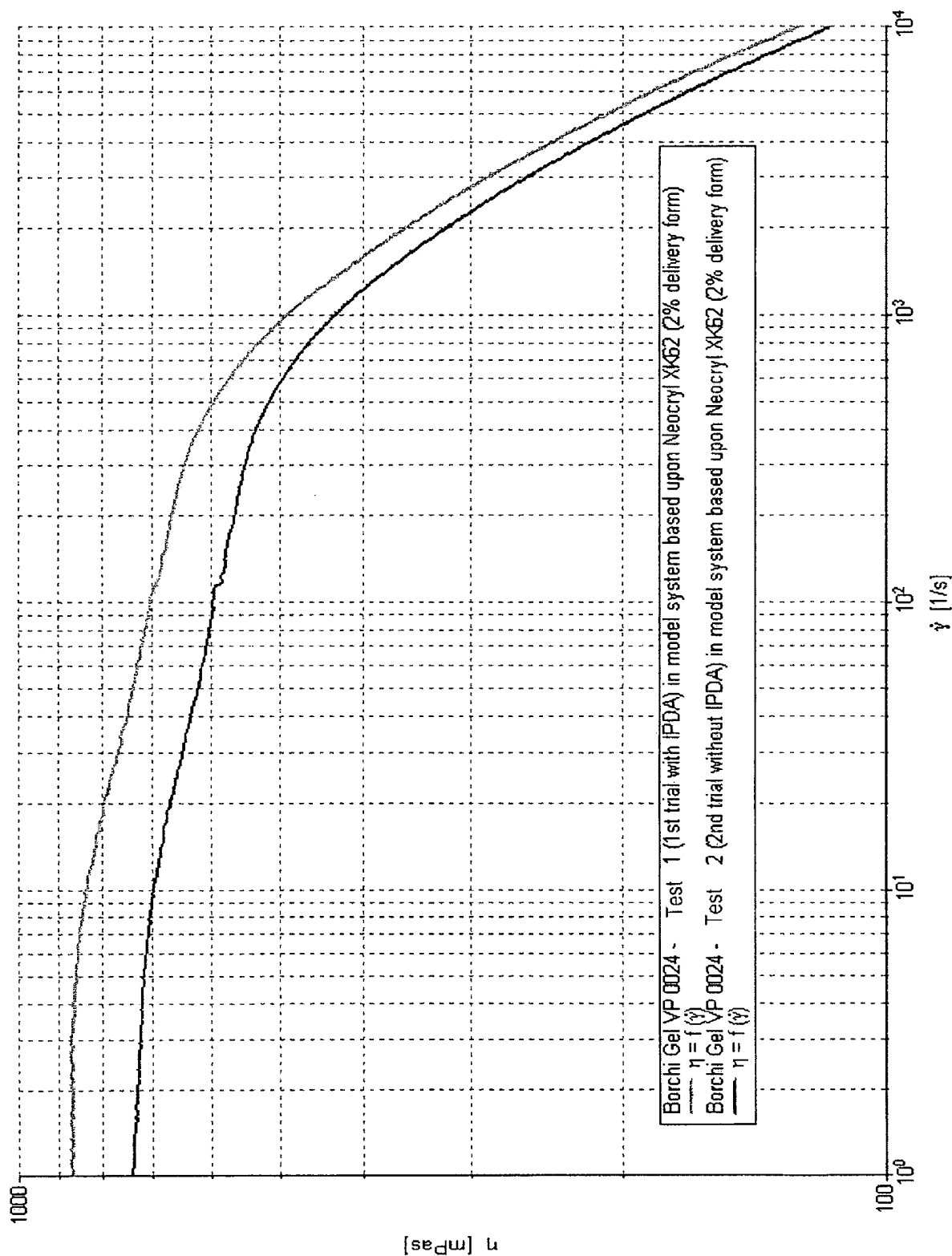
imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statement may jeopardize the validity of pending Application Ser. No. 10/092,212 or any patent issuing thereon.

Signed at Langenfeld , this 19 day of November , 2004.

A handwritten signature in cursive script, appearing to read "Frank Sauer", written over a horizontal dashed line.

(Frank Sauer)

Appendix 1: Comparative trials with Borch Gel VP 0024 (with and without use of isophorone diamine (IPDA))



Appendix 2: Comparative trials based upon example 74M in Emmons patent 4,079,028

